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INFLUENCE OF THE ANIONIC COMPOSITION AND ACID GAS CONTENT IN FORMATION WATERS OF PERM KRAI FIELDS ON CORROSION INHIBITOR EFFECTIVENESS

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ВЛИЯНИЕ АНИОННОГО СОСТАВА И СОДЕРЖАНИЯ КИСЛЫХ ГАЗОВ В ПЛАСТОВЫХ ВОДАХ МЕСТОРОЖДЕНИЙ ПЕРМСКОГО КРАЯ НА ЭФФЕКТИВНОСТЬ ИНГИБИТОРОВ КОРРОЗИИ

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In the course of hydrocarbon deposit development, formation water starts entering the wells over time. An increasing produced water volume has a negative impact on economic indicators of oilfield developments and leads to other hazardous consequences, such as equipment corrosion. Downhole equipment corrosion causes largest financial losses, since well accidents entail expensive repairs. Therefore, it is critical to prevent the equipment corrosion. The study considers main properties and composition of formation waters. Based on an overview of open literature sources, we analyzed the impact of Cl^- , SO_4^{2-} , HCO_3^- anions contained in the formation waters on corrosion process kinetics at the phase interface between metal (electrode) and water (electrolyte). The study also discusses the results of researches on the influence of dissolved acid gases (H_2S and CO_2) on corrosion mechanisms and rates. An influence of pH value on the corrosion rate is considered. Contents of various salts in the formation waters contribute to the complexity of corrosion processes occurring in real conditions. Presence of aggressive environments can accelerate corrosion in some instances, but sometimes it can slow it down; this fact also affects the inhibitor protection effectiveness.

Based on laboratory analysis data, effectiveness of using the corrosion inhibitor in the formation waters with different compositions and properties is assessed. It has been established that presence of a mix of aggressive components – chloride ions and hydrogen sulfide – in the formation waters causes hydrogen sulfide corrosion inhibition and reduction of chloride corrosion rate. Presence of dissolved salt ions and acid gases in the formation waters improves effectiveness of the corrosion inhibitors due to strong passive films formed on steel equipment surfaces.

Ключевые слова:

питтинговая коррозия, скважина, сероводород, хлориды, сульфаты, бикарбонаты, диоксид углерода, двойной электрический слой, пассивация, потенциал питтингообразования, вольтамперометрия, импедансная спектроскопия, анионы, катионы, ингибитор коррозии.

При разработке залежей углеводородов в скважины со временем начинают поступать пластовые воды. Повышение объемов попутно добываемых вод негативно сказывается не только на экономических показателях разработки месторождений, но и приводит к другим опасным последствиям, к таким, например, как коррозия оборудования. Наибольший экономический ущерб приносит коррозия скважинного оборудования, так как аварии на скважинах обуславливают необходимость проведения дорогостоящих ремонтных операций. В связи с этим предотвращение коррозии оборудования является актуальной задачей. Рассматриваются основные свойства и состав пластовых вод. На основании открытых литературных источников анализируется влияние содержащихся в пластовых водах анионов Cl^- , SO_4^{2-} , HCO_3^- на кинетику протекания коррозионных процессов на поверхности раздела фаз металла (электрода) и воды (электролита). Также приведены данные исследований влияния наличия растворенных кислых газов (H_2S и CO_2) на механизм и скорость протекания коррозии. Рассматривается влияние водородного показателя на скорость коррозии. Содержание различных солей в пластовых водах обуславливает сложность коррозионных процессов, происходящих в реальных условиях. Наличие агрессивных сред в некоторых случаях способствует ускорению коррозии, но иногда и ее замедлению, это также сказывается на эффективности ингибиторной защиты.

На основе данных лабораторных исследований проведена оценка эффективности применения ингибиторов коррозии в пластовых водах с различными составами и свойствами. Установлено, что содержание в пластовой воде смеси агрессивных компонентов – хлорид ионов и сероводорода – приводит к угнетению сероводородной коррозии, при этом скорость хлоридной коррозии тоже снижается. Наличие ионов растворенных солей и кислых газов в пластовых водах способствует повышению эффективности применения ингибиторов коррозии благодаря образованию прочных пассивных пленок на поверхности стального оборудования.

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Introduction

During operations of producing wells, oil outputs decrease and volumes of produced formation water increase. Exposure of the downhole equipment to the formation water causes its intense corrosion wear. Equipment corrosion is also influenced by presence of acid gases (hydrogen sulfide and carbon dioxide) in the produced oil. Corrosion reduces mean time between failure of the downhole oil producing equipment and leads to high financial and environmental losses. Chemical equipment protection methods are widely used nowadays; they are based on regulating the electrochemical balance in liquid environments or formation of protective films on metal surfaces of the equipment. A high watercut of wellbore fluids makes chemical protection with corrosion inhibitors very expensive.

Formation Water Composition

Formation waters of hydrocarbon fields are balanced thermodynamic systems that consist of solutions of various salts and gases. Ionic composition of the produced water in the hydrocarbon fields of Perm Krai is mostly represented by cations (Na^+ , K^+ , Ca^{+2} , Mg^{+2}) and anions (Cl^- , SO_4^{2-} , HCO_3^-). Depending on the genesis of the formation waters, a quantitative content of each ion can vary. Formation waters of hydrocarbon fields typically have high content of Cl^- ions – up to 300 g/L; the origin of chloride ions is salt solutions (KCl , NaCl , CaCl_2 , MgCl_2 etc.). The presence of sulfates and bicarbonates is caused by dissolution of carbonate rock (CaCO_3 and MgCO_3) and gypsum (CaSO_4); the content of these anions changes depending on a certain formation water type and usually does not exceed 3 g/L.

Apart from salts, formation waters can also contain dissolved acid gases – H_2S и CO_2 , originating from an associated occurrence of hydrocarbons with formation waters. Properties of formation waters also depend on pH value which can range from 5 to 8 in natural conditions of oil and gas formations occurrence. Presence of each component in the formation waters determines its type and corrosive effect on metals. A detailed analysis of influence of the formation water composition on a metal corrosion rate is provided below.

Influence of Formation Water Ionic Composition on Steel Corrosion Rate

Corrosion of metals in liquid environments containing dissolved salts has an electrochemical mechanism and occurs at the interface between a metal

(electrode) and electrolyte (formation water). It involves formation of adsorption films on metal surfaces – double electric layers (DEL), mostly represented by anions of salts dissolved in electrolytes. The double electric layer at the electrode-electrolyte interface possesses electric resistance and a certain electric capacity corresponding to its molecular thickness. DEL electric properties characterize the degree of chemical interaction between electrode and electrolyte and are used to determine metal corrosion rate employing such methods as, e.g., impedance spectroscopy and voltammetry. Impedance spectroscopy methods consist in supply of a sinusoidal low amplitude excitation signal to the system under research and studying the output response signal [1]. Voltammetry is a method that captures changes over time of the current flowing through the system under study when exposed to voltage [2]. The higher is DEL electrical conductivity, the faster are surface chemical reactions, including metal corrosion. The more is DEL thickness and the less its electrical conductivity, the thicker and the stronger is the passivation film and the lower is the corrosion rate.

The most hazardous type of steel equipment corrosion in oil and gas wells is pitting corrosion. It causes local cavitation of metal, which can lead to premature failure of equipment, while most of the metal surface can still remain intact.

Occurrence and progress of pitting corrosion on metal surfaces in electrolytes occurs when the critical voltage potential of pitting corrosion formation is exceeded. Presence of aggressive anions in electrolytes (normally chlorides) facilitates reduction of pitting corrosion critical potential and acceleration of corrosion processes [3]. In absence of chloride ions, oxide film of stainless steel 304 remains passive until achievement of values of oxygen release voltage potential (+1.2 V). In NaCl solution with the concentration of 0.1 mole, pitting starts to occur at the voltage potential of +0.35 V (Fig. 1). In Na_2SO_4 solution with the concentration of 1 mole, pitting does not occur, which confirms the fact that pitting corrosion is mostly caused by presence of chloride ions [4].

As the content of chloride ions in electrolytes increases, pitting corrosion voltage potential becomes significantly lower and steel corrosion accelerates (Fig. 2) [5].

There are many mechanisms of pitting occurrence, with the following basic features [6–12]:

- 1) at the initial stage, anions from electrolytes concentrate on the passive film surface
- 2) at the following stage, anions destroy or dissolve the passive film
- 3) after initiation, current density in the pitting growth focus significantly increases due to relatively small size of pitting surface vs. intact metal surface.

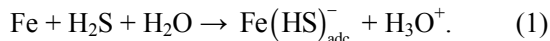
As the pitting and its volume grow, cations of dissolved metal remain inside it and don't diffuse out from porous products of corrosion. As a result, the accumulated metal cations undergo hydrolysis and acidity grows inside the pitting (Fig. 3, a). Besides, electrode potential above the pitting is more active than on the adjacent surface (Fig. 3, b) [13].

Thus, occurrence of pitting corrosion includes formation of acid corrosive internal electrolyte. The system that is formed is stable and stimulates constant further growth of pitting.

Presence of sulfate ions increases pitting corrosion voltage potential, which slows it down [14–17] (Fig. 4).

HCO₃⁻ anion facilitates deceleration of corrosion processes through interaction with calcium cations and forming calcium bicarbonate Ca(HCO₃)₂. Calcium bicarbonate is the cheapest cathode screening inhibitor of steel corrosion and, owing to formation of insoluble calcium carbonate which precipitates on metal surface, is used in water supply systems.

Hydrogen sulfide H₂S is an aggressive gas which dissolves in water and forms weak acid that causes corrosion of metals with formation of sulfides, which play the role of cathode in regard to iron and form a galvanic pair with it, which leads to quick deterioration of metal structures [18, 19]. Hydrogen sulfide speeds up the anode reaction of iron ionization:



In simultaneous presence of hydrogen sulfide and acids in the solution, corrosivity of both components increases drastically [20, 21]. Corrosion rate in 1N HCl solution sharply increases as the temperature rises. Hydrogen sulfide mostly accelerates anode process of corrosion than the cathode process [22] due to formation of a surface compound (FeHS)_{ads}⁻ that is a catalyst of anode reaction of metal ionization [23–28].

Carbon and low-alloy steels that are most commonly used in construction of oil and gas pipelines are prone to corrosion in CO₂ containing environments. Carbon dioxide solution in water phase forms carbonic acid which causes steel corrosion:



The mechanism of iron dissolution in carbonic acid [29] is represented by cathode (3), (4), (5) and anode (6) reactions. At ambient pH equal to 6, H₂CO₃ (2) and HCO₃⁻ decompose (3).

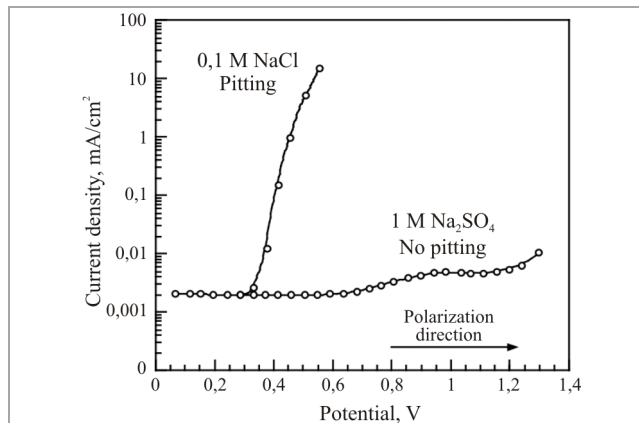
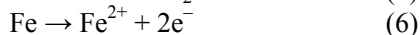
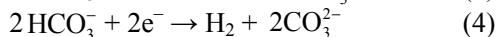
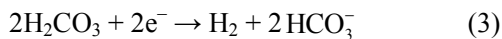


Fig. 1. Polarization curves for stainless steel in 0.1 mole NaCl solution and 1 mole Na₂SO₄ solution [4]

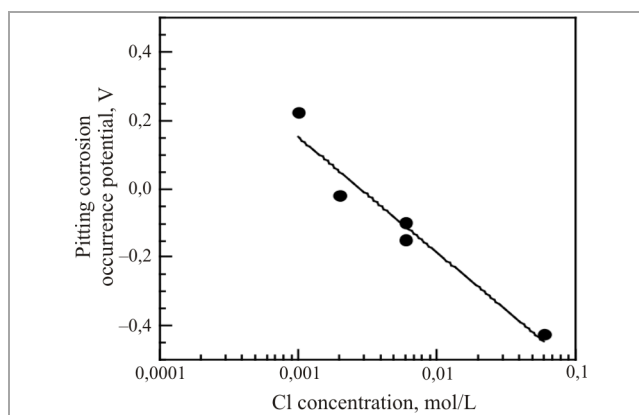


Fig. 2. Impact of chloride ions content on pitting corrosion occurrence potential [5]

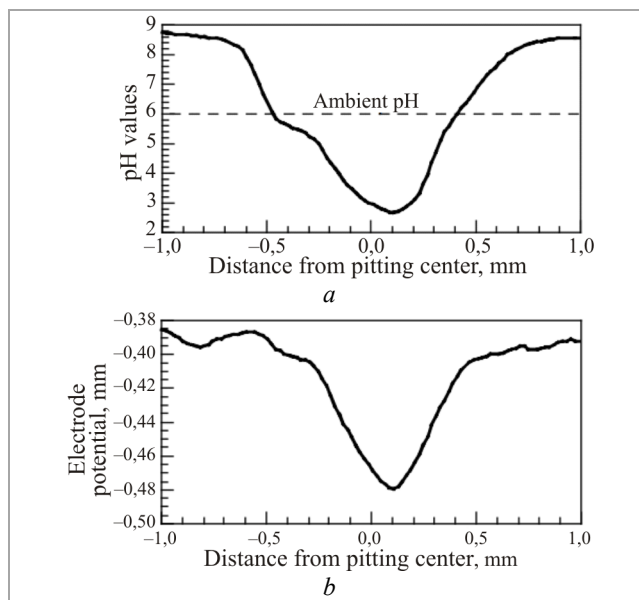
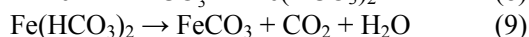
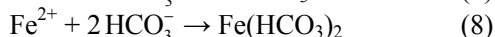


Fig. 3. Change of pH (a) and electrode potential (b) against growing pitting on steel surface in weak NaCl solution [13]

As a result of these processes, a corrosive layer forms on steel surface. Properties of this layer impact steel corrosion rate in CO₂ water solutions. Some of the studies suggest that low-soluble iron carbonate sediment FeCO₃ can form protective layers [29–34], which is explained by equations (7), (8) and (9) [35, 36]:



However, the protective layer formed by FeCO₃ sediment is porous and fragile, and only slightly reduces corrosion rate due to the reduction of mass transfer between corrosive medium and clean surface of steel [37].

If FeCO₃ protective film forms on steel surface, then emergence of the second capacity arch on Nyquist plot or second time constant on Bode plot can be expected [37]. However, this is not observed on impedance plots (Fig. 5) [38]. This is due to formation of FeCO₃ porous thin layer with resistance far lower than charge transfer resistance [39, 40].

Increase of capacity values over time (loop diameter increase) is related to the increase in iron carbonate sedimentation area on sample surface, which is accompanied by increasing charge transfer resistance [41].

Ambient pH influence has been studied in [42], which concluded that decrease of ambient pH from 8 to 2 causes a reduction of charge transfer resistance (R_{ct}) by more than two times, which suggests an increase in corrosion rate in acid environments (Fig. 6).

The content of various salts in formation waters contributes to the complexity of corrosion processes occurring in real conditions. Presence of aggressive environments can accelerate corrosion in some instances, but sometimes can slow it down. The conducted research of steel corrosion in formation water analogue with various levels of chloride and sulfate ions and hydrogen sulfide led to the following conclusions [43]:

- presence of sulfate ions in electrolytes facilitates steel surface passivation, whereas hydrogen sulfide content up to 150 mg/L improves passivation, which is caused by formation of two different passivation products (iron oxides and sulfides) that inhibit anode oxidation of steel;

- content of chloride ions in water from 1 to 40 g/L and hydrogen sulfide up to 150 mg/L does not reduce corrosion rate;

- at the levels of content of chloride ions in water from 160 g/L, presence of hydrogen sulfide does not contribute to development of corrosion, which is caused by prevalence of chloride ions in corrosion reactions.

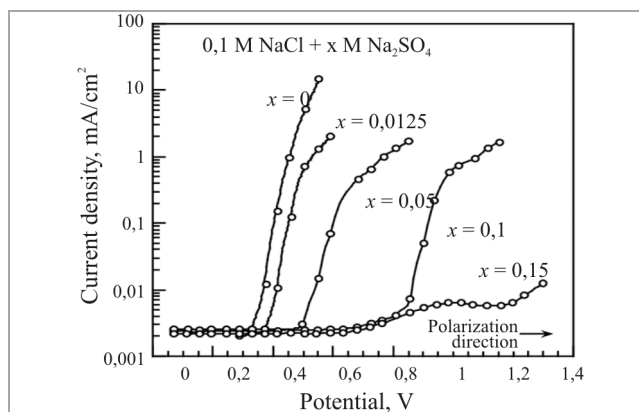


Fig. 4. Impact of sodium sulfate concentration on pitting corrosion occurrence potential on stainless steel 3004 in NaCl solution

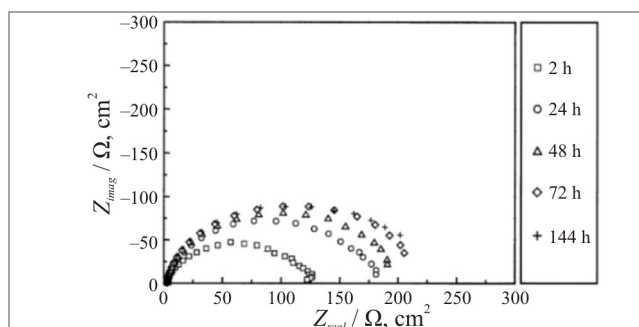


Fig. 5. Impedance diagram for steel after curing in CO₂ solution during 2, 24, 48, 72 and 144 h [38]

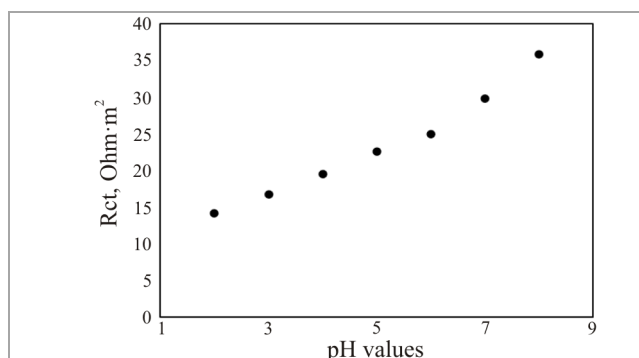


Fig. 6. Impact of ambient pH on charge transfer resistance R_{ct} on steel surface in 1N HCl solution

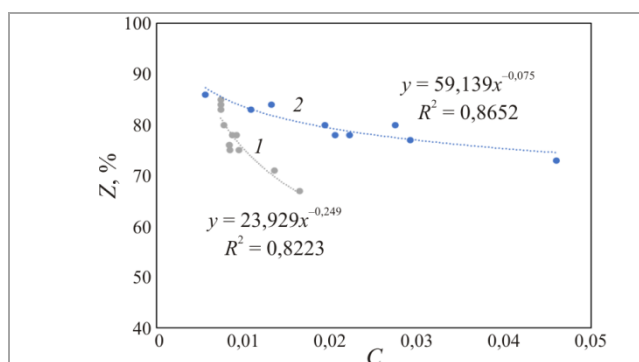


Fig. 7. Impact of anion content on effectiveness of corrosion inhibitors: 1 – Cl⁻ content more than 60,000 mg/L; 2 – Cl⁻ content less than 60,000 mg/L

Results of research of corrosion inhibitor effectiveness and formation water properties in Perm Krai fields

Sr. No.	Well	Effectiveness, %	H ₂ S, mg/L	CO ₂ , mg/L	pH	ρ, g/cm ³	Total salinity, g/L	HCO ₃ ⁻ , mg/L	SO ₄ ²⁻ , mg/L	Cl ⁻ , mg/L	Ca ²⁺ , mg/L	Mg ²⁺ , mg/L	K ⁺ + Na ⁺ , mg/L	C
Group 1														
1	934	75	151.60	35	6.71	1.098	143.80	210	625	88300	10200	2580	41885	0.0095
2	2606	78	112.30	38	6.56	1.105	155.65	190	690	96000	10200	2700	45873	0.0092
3	1299	80	356.80	30	6.19	1.095	145.80	183	509	89900	9900	2260	43003	0.0077
4	797	83	118.50	30	6.19	1.115	165.04	170	580	102000	10800	2950	48544	0.0074
5	868	84	98.50	30	6.46	1.114	165.04	170	580	102000	10800	2950	48544	0.0074
6	922	85	138.00	33	7.00	1.075	108.80	270	230	67600	6830	2300	31906	0.0074
7	193	76	132.00	35	6.11	1.070	103.90	305	230	64230	6140	2078	30900	0.0083
8	905	67	206.30	35	6.23	1.108	156.40	140	1440	96000	10200	2950	45740	0.0165
9	498	71	132.40	50	6.58	1.115	165.00	170	580	102000	10800	2950	48544	0.0074
10	241	75	58.70	30	6.21	1.131	193.20	122	888	120000	14700	3040	54228	0.0084
11	193	76	132.00	35	6.11	1.070	103.90	305	230	64230	6140	2078	30900	0.0083
Group 2														
1	953	73	98.00	35	7.32	1.057	84.02	390	1950	50900	6060	1500	23220	0.0460
2	2264	77	164.10	30	6.54	1.058	77.02	390	950	45900	5060	1500	23216	0.0292
3	229	80	4.30	35	7.00	1.049	73.33	545	290	43100	5083	1220	24216	0.0194
4	335	78	0.00	50	6.58	1.021	35.60	240	298	24283	2050	450	8240	0.0222
5	542	78	53.00	15	7.06	1.046	68.30	250	594	41122	4609	1093	20556	0.0205
6	508	83	57.00	27	7.19	1.060	95.30	162	458	57105	6679	1516	29126	0.0109
7	563	84	137.80	45	7.18	1.067	96.60	200	589	59900	6500	1700	27711	0.0132
8	859	86	67.80	35	7.36	1.066	95.90	210	123	59600	6570	1800	27883	0.0056
9	1026	80	75.90	47	6.45	1.062	97.40	326	1290	58900	4610	1580	30676	0.0274

Laboratory Data Analysis

Use of corrosion inhibitors helps slow down electrochemical reactions on steel surfaces. Corrosion inhibitor application effectiveness mostly depends on electrolyte content. In certain instances, presence of dissolved salt ions improves inhibitor effectiveness due to formation of strong passive films on steel equipment surfaces [44, 45].

Effectiveness of imidazoline corrosion inhibitor used in Perm Krai oilfields was determined in laboratory conditions using cyclic voltammetry (CVA). Corrosion inhibitor was added to formation water from Perm Krai fields in the concentration of 100 g/m³. Research results and formation water content are shown in the table (table). The research data has been split into two groups based on chloride ions content in formation waters.

Corrosion inhibitor effectiveness (Z) was determined from the following ratio:

$$Z = \frac{I_o + I_{in}}{I_o}, \quad (10)$$

where I_o and I_{in} – maximum anode current densities before and after addition of inhibitor, respectively.

Laboratory analysis has shown that effectiveness of imidazoline corrosion inhibitor is mostly influenced by anion content ratio (HCO₃⁻, SO₄²⁻ and Cl⁻) in formation water (Fig. 7):

$$C = \frac{C_{\text{HCO}_3^-} + C_{\text{SO}_4^{2-}}}{C_{\text{Cl}^-}}, \quad (11)$$

where $C_{\text{HCO}_3^-}$, $C_{\text{SO}_4^{2-}}$, C_{Cl^-} – weight content of bicarbonate ions, sulfate ions and chloride ions in formation water, respectively.

Based on the results of laboratory analysis, it has been established that at high content of chloride ions, effectiveness of imidazoline corrosion inhibitor mostly depends on the content of bicarbonate and sulfate ions (Fig. 7, curve 1). No evidence of impact of hydrogen sulfide content on inhibitor effectiveness has been obtained, which can be caused by prevalence of chloride ions. For the second group, with lower content of chloride ions, dependence of inhibitor effectiveness on C is more flat; impact of hydrogen sulfide was not observed either.

Conclusion

Corrosion activity of formation waters in Perm Krai fields is caused by presence of aggressive substances (Cl⁻, SO₄²⁻, HCO₃⁻, H₂S and CO₂), each of them having an individual impact on corrosion acceleration. Presence of a mix of aggressive components (chloride ions and hydrogen sulfide) in formation water of Perm Krai fields leads to inhibition of hydrogen sulfide corrosion; chloride corrosion rate also goes down.

Presence of anions of dissolved salts (Cl⁻, SO₄²⁻, HCO₃⁻) and acid gases (H₂S, CO₂) in formation waters

improves effectiveness of imidazoline corrosion inhibitor in Perm Krai fields owing to formation of strong passive films on steel equipment surfaces.

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